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Cicerostr. 24
D-10709 Berlin
Germany
Tel +49 (0)30 536 53 800
Fax +49 (0)30 536 53 888
www.kompetenz-wasser.de

Extended summary

Optimization of flocculation for tertiary filtration
processes and evaluation of sustainability of
tertiary wastewater treatment

Project acronym: OXERAM 2

by
Ulf Miehe, Johan Stüber, Christian Remy, Margarethe Langer
Manuel Godehardt, Morgane Boulestreau

Kompetenzzentrum Wasser Berlin gGmbH

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Colophon

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Title

Extended summary: Optimization of flocculation for tertiary filtration processes and evaluation of sustainability of tertiary wastewater treatment

Authors

Ulf Miehe, Kompetenzzentrum Wasser Berlin gGmbH

Morgane Boulestreau, Kompetenzzentrum Wasser Berlin gGmbH

Margarethe Langer, Kompetenzzentrum Wasser Berlin gGmbH

Christian Remy, Kompetenzzentrum Wasser Berlin gGmbH

Manuel Godehardt, FG Wasserreinigung, TU Berlin

Johan Stüber, Kompetenzzentrum Wasser Berlin gGmbH

Quality control

Boris Lesjean, Kompetenzzentrum Wasser Berlin gGmbH

Daniel Mutz, Kompetenzzentrum Wasser Berlin gGmbH

Publication / Dissemination approved by technical committee members:

C. Bourdon, Veolia

A. Tazi-Pain, Veolia

C. Bartholomäus, Berliner Wasserbetriebe

R. Gnirß, Berliner Wasserbetriebe

A. Peter-Fröhlich, Berliner Wasserbetriebe

M. Jekel, FG Wasserreinigung, TU Berlin

A. Hartmann, Kompetenzzentrum Wasser Berlin gGmbH

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Introduction

OXERAM 2 was a long term project which included the assessment of pre-ozonation as pre-treatment to reduce membrane fouling and the assessment of the micro sieve technology for advanced phosphorus removal. Additionally the sustainability of different processes for advanced wastewater treatment such as rapid filtration, membrane filtration, UV disinfection, adsorption and ozonation was evaluated.

Pilot scale installations for ozonation, polymeric membrane, ceramic membrane, and micro sieve were operated in parallel at WWTP Ruhleben quantifying the key parameters for LCA/LCC assessment. In order to understand the fundamental mechanisms of membrane fouling influenced by coagulation and/or ozonation lab scale investigations were carried out at TU Berlin, Chair of Water Quality Control.

The present summary collects the abstracts and key graphs of each detailed report. The following detailed reports are listed below and can be downloaded here:

<http://www.kompetenz-wasser.de>.

- Tertiary treatment combining ozonation and membrane filtration - Pilot scale investigations
- Guidelines for the use of online fouling monitoring in tertiary treatment
- Role of organic substances in tertiary treatment via oxidation and membrane filtration
- Feasibility of the microsieve technology for advanced phosphorus removal
- Life Cycle Assessment and Life Cycle Costing of tertiary treatment schemes

These reports represent the previous mentioned work packages. The following table summarizes the different process options and costs for the extension of WWTP Ruhleben.

Criteria		Actiflo™ + UV	Microsieve (10 µm) + UV	Dual media filter + UV	Polymeric UF + UV-bypass	Ceramic MF + UV-bypass
Data basis		Manufacturer information + modeling	Pilot trials OXERAM	Pilot trials BWB	Pilot trials OXERAM	Pilot trials OXERAM
		Optimized design based on pilot scale trials				
Design flow	[m³/s]	7.4	7.4	7.4	4.5 (+UV) ³	4.5 (+UV) ³
TP	[µg/L TP _{85%}]	< 120 - 150	< 80	< 80	< 50	< 50
	[µg/L TP _{mean value}]	90 - 120	60 - 65	50 - 60 ⁵	20 - 25	20 - 25
	Load removal [t P/a]	- 18.6	- 22.8	- 23.4	- 25.4	- 25.4
SS	[mg/L _{85%}]	< 7	< 3	< 1	<< 1	<< 1
Capital cost	[Mio. €]	30.4	37.0	41.5	65.2	95.4
Operational cost	[ct/m³]	3.1	3.3	2.9	5.8	7.9
Annual cost (CAPEX + OPEX)	[ct/m³]	5.8	6.5	6.5	11.7	13.9
	[€/kg P _{elim}]	261 - 287	246-251	239 - 248	399-406	471-479
Energy demand (filtration/ sedimentation)	[Wh/m³]	31	35	42	88	88
Design UV disinfection	[J/m²]	850	700	700	1000 ³	1000 ³
CO ₂ footprint	[kg CO ₂ -eq/ kg P _{elim}]	224 - 246	177-180	185 - 192	275 - 280	272-276
Return flow	[%]	4	1.8	4 - 5	5 ¹	5
Extension to micro-pollutant removal	<i>Powdered activated carbon</i>	Yes (PAC circulation + additional filtration)	uncertain (PAC choice complex)	Yes (GAC potentially possible)	Yes (primarily tested in water production)	? (in theory possible)
	<i>Ozonation</i>	? ²	? ²	Yes	? ²	? ²
Full-scale application	<i>For above mentioned TP goal</i>	Yes	No ⁴	No ⁶	Yes	No
	<i>Generally</i>	> 100	> 50 (without coagulation)	> 100	> 50	1
¹ Recovery in pilot trials > 94 % ² For ozonation the necessity of a biological activated fixed bed filter as post-treatment is discussed but hitherto not mandatory ³ The amount of water > 4.5 m³/s is treated via UV disinfection. ⁴ For TP < 150/100 three plants are under construction in Europe and two in the US ⁵ Result "Raumfiltration" (BWB): 60 µg/L TP WWTP Münchehofe; Results "IST4R" and "ASKURIS": 53 µg/L TP WWTP Ruhleben (mean values) ⁶ Target concentration hitherto only achieved with processes including 2-stage flocculation and sedimentation (e.g. SWTP Tegel)						

Tertiary treatment combining ozonation and membrane filtration Pilot scale investigations

Within the project OXERAM state of the art membrane filtration was applied as a tertiary treatment step for advanced phosphorus removal in a municipal wastewater treatment plant. Two membrane types, ceramic and polymeric, were tested in pilot scale (up to 3 m³/h per module), using commercial membrane modules. Due to the drawback of membrane fouling, leading to comparably high investment and operating costs, pre-treatment with ozone was tested. Ozonation was expected to increase the sustainable flux for both membrane types.

For both membrane types high filtrate quality was achieved. A mean total phosphorus concentration below 25 µg/L was achieved over two years. Additionally disinfection is reached and therefore the European bathing water standards were met.

Ultrafiltration modules (0.02 µm) made of polyether sulfone (PES) and delivered by company inge (Germany) were tested comparing different capillary diameters (0.9 vs. 1.5 mm) leading to different package densities (respectively 40 and 60 m² per module). Both types were operated in parallel and the experience showed a more robust operation with 1.5 mm capillaries when applying high fluxes targeting high recoveries. Both evaluation parameters, total fouling rate and membrane regeneration by cleaning in place, suggested the 1.5 mm module for the application at the WWTP Ruhleben. Optimizing the operation set up and cleaning strategy proved that recoveries ≥ 95 % could be achieved and therefore a second filtration stage treating the backwash water is obsolete. The design with max 75 L/(m²h), 60 minutes of filtration, and a backwash duration of 40 s is the proposed set up for WWTP Ruhleben. A daily acidic chemical enhanced backwash combined with a weekly caustic cleaning step proved to manage the fouling affinity and a cleaning in place interval of 1 – 3 months was demonstrated in a long term run, see Figure 1. The usage of ozone did not improve the overall filtration performance, because the benefit of a higher filterability is compensated by a higher additional fouling resistance after each backwash. Therefore the mean trans-membrane pressure remained in the same range. These results were only observed with the combination of ozonation and PES ultrafiltration membranes. Lab scale tests conducted at the Chair of Water Quality, TU Berlin, confirm this outcome but showed different results for other membrane materials and pore sizes.

The potential to reduce the total fouling rate combining ozonation with coagulation prior ceramic membrane filtration was shown. A microfiltration membrane (0.1 µm monolith module provided by company NGK, Japan) consisting of Al₂O₃ and a surface of 25 m² was tested in pilot scale. Applying a dose of 15 mgO₃/L ($z = 1.18 \text{ mgO}_3/\text{mgDOC}$) could reduce the total fouling rate by half even when doubling the flux from 60 L/(m²h) to 120 L/(m²h). Critical flux experiments showed that the application of 7.5 mgO₃/L ($z = 0.7 \text{ mgO}_3/\text{mgDOC}$) was sufficient to achieve the beneficial effect of pre-ozonation. Treating the secondary effluent of WWTP Ruhleben a sustainable flux around 130 – 140 L/(m²h) was identified when applying pre-ozonation of 7.5 mgO₃/L ($z = 0.7 \text{ mgO}_3/\text{mgDOC}$) and 8 mgFe/L for coagulation. It was not possible to demonstrate this process set up in a long term run, due to technical malfunctions. However, an economic evaluation showed that for the case of WWTP Ruhleben a sustainable flux > 500 L/(m²h) is required to be competitive against tertiary treatment with polymeric membranes without ozone. This high value can be explained by the high module cost for ceramic membranes and the high DOC content of the secondary effluent, leading to increased effort for ozonation.

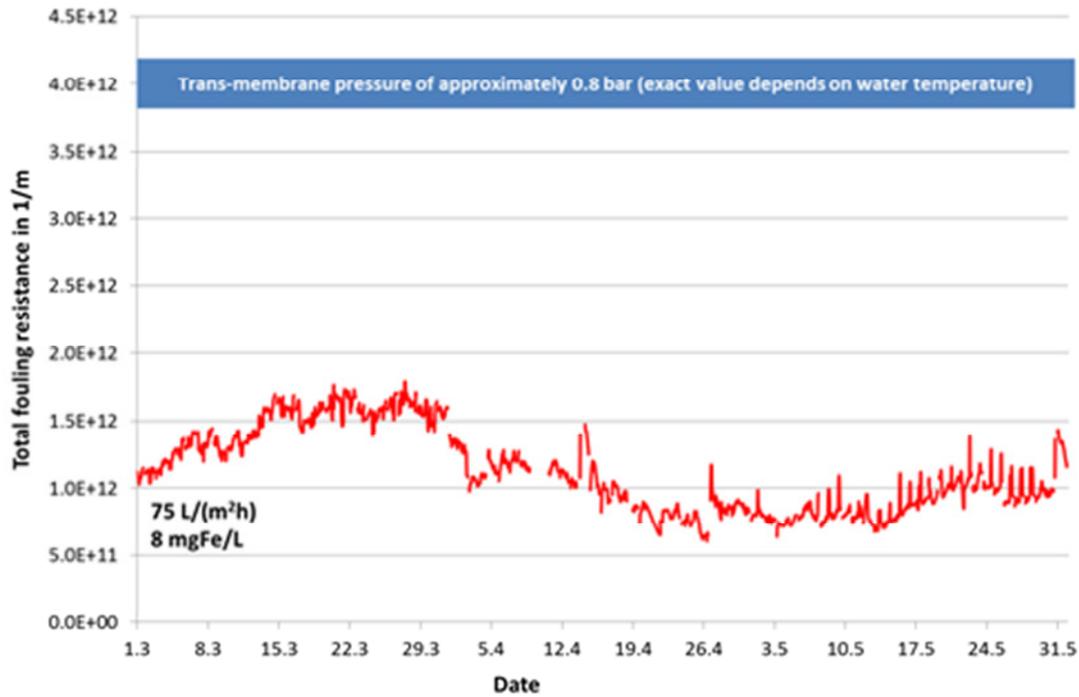


Figure 1: Long term demonstration - Fe as coagulant

The identified operation parameters were demonstrated in an uninterrupted operation lasting more than 3 months. This demonstration phase shows the feasibility of polymeric membranes for tertiary treatment processes achieving high recoveries (~95 %) and total phosphorus effluent concentrations <<50 µg/L.

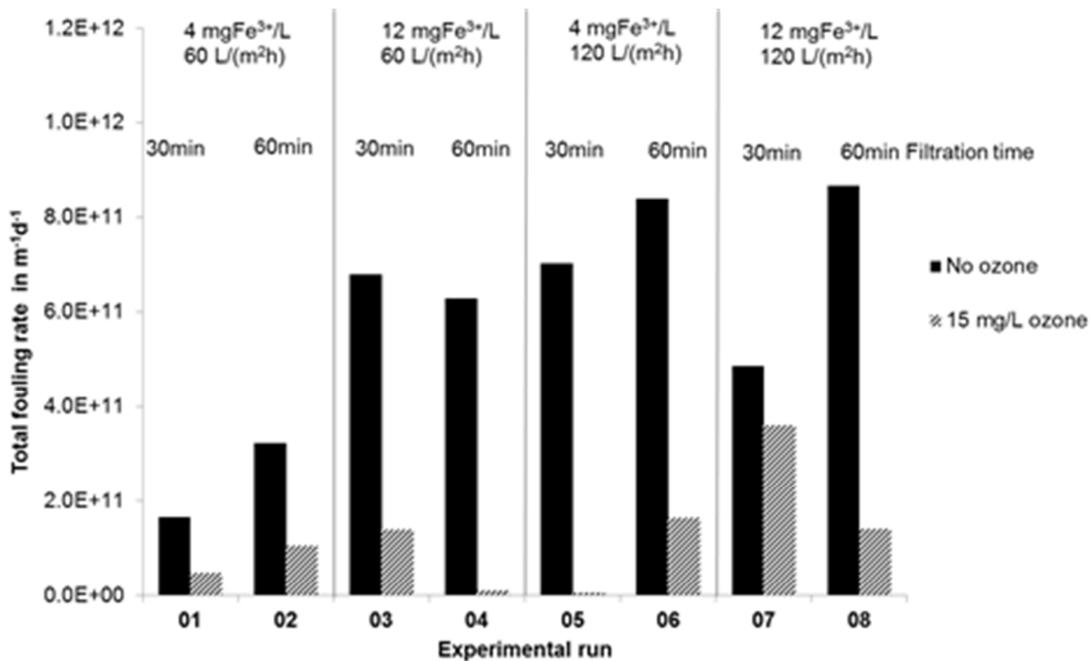


Figure 2: Total fouling rates ceramic membrane - trial phase 1

Pre-ozonation reduces the total fouling rate, see Figure 2, of the subsequent microfiltration step using a ceramic membrane module. Therefore higher fluxes can be applied and cleaning needs are reduced minimizing the operational costs of the membrane filtration step.

Guidelines for the use of online fouling monitoring in tertiary treatment

Various tertiary treatment processes were compared in the OXERAM project, including a polymeric membrane and a microsieve pilot plant which were installed at the Ruhleben WWTP in Berlin and operated for almost two years. To increase the performance of both processes, pre-treatment with ozonation, coagulation and/or flocculation were tested. In order to optimize the hybrid processes and to develop a control strategy, online monitoring was implemented. After a literature review and lab trials at the Chair of Water Quality, TU Berlin during the project preparation phase, two instruments were recommended.

One NS500 device by Nanosight (UK) was installed at the influent of the UF membrane pilot (pore diameter = 20 nm) influent with sampling every 15 minutes before and after the inline coagulation. The particles between 50 and 1000 nm were analysed to evaluate the impact of the ozonation / coagulation or the coagulation alone on the nanoparticles below 500 nm which are most responsible for fouling. For a better reproducibility and quality of the results, samples were pre-filtered by an online metallic 5 µm filter. Particle analysis by Nanoparticle Tracking Analysis (NTA) was obtained to give reliable and reproducible information about the concentration and size distributions of the colloidal fraction in the tested treated domestic wastewater. Correlation between the membrane reversible fouling measured with the help of the trans-membrane pressure (TMP) and the concentration of particles between 100 and 200 nm were detected. Online measurements at the pilot-scale indicate that colloid peak concentrations can be compensated for by coagulation with an optimum dose of 8 mg Fe³⁺/L. Furthermore, a comparison of FeCl₃ and PACl demonstrated that the former is more effective in colloid removal for this treated domestic wastewater. Due to the combination of pre-ozonation and subsequent coagulation, a synergy effect was determined as the combined treatments lead to a better particle removal compared to the effect of the single treatments at same dosages of O₃ and Fe³⁺. A combination of 0.5 mg O₃/mg DOC₀ and 8 mg Fe³⁺/L leads to a total reduction down to < 5 % of the initial colloid content, see Figure 3. However, a direct prediction of irreversible fouling was not possible. This device should be further optimized for its potential to reduce operational costs and lower solid loads and thus fouling on the membrane.

A particle counter device by Pamas (Germany) was installed in the 10 µm mesh size microsieve effluent pipe bypass and this measured the particle size distribution continuously by light extinction at a wavelength of 635 nm at 25 mL/min. No pre-treatment was necessary and it was possible to automatically clean the instrument every hour with distilled water or another cleaning solution. Piping and sensor cell maintenance was crucial to improve the quality of the results due to the high potential of the effluent water to post-flocculate. For optimization of the coagulant and flocculant mixing velocity, the particle counter results were more accurate than the turbidity sensor which did not detect any changes in the effluent water quality. The monitoring tool detected the lowest particle concentration for the optimized mixing velocity, see Figure 4. However, the particle counter did not provide better information than an online turbidity sensor for other parameters such as the coagulant types or doses. Therefore, while it is recommended to use an online particle counter during the microsieve plant start-up phase to optimize the coagulation and flocculation, for routine controls an online turbidity sensor is sufficient. Moreover turbidity sensors are less demanding in terms of maintenance effort. The project showed that using the turbidity signal on raw water to adapt the coagulant dose was very efficient.

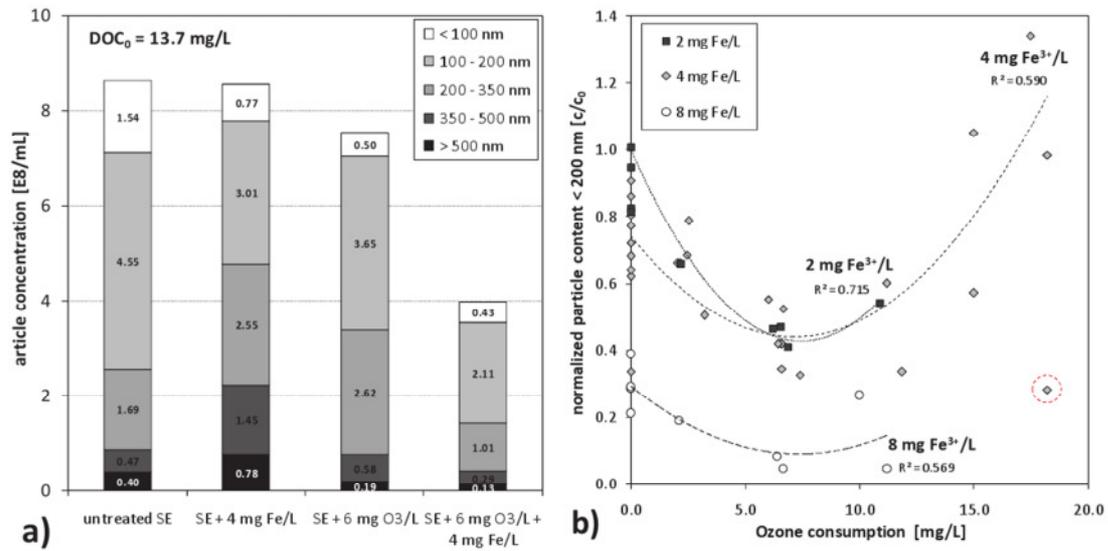


Figure 3: Impact of pre ozonation and subsequent coagulation on the submicron particle content a) size distribution, b) particle removal < 200 nm at different ozone and coagulant dosages

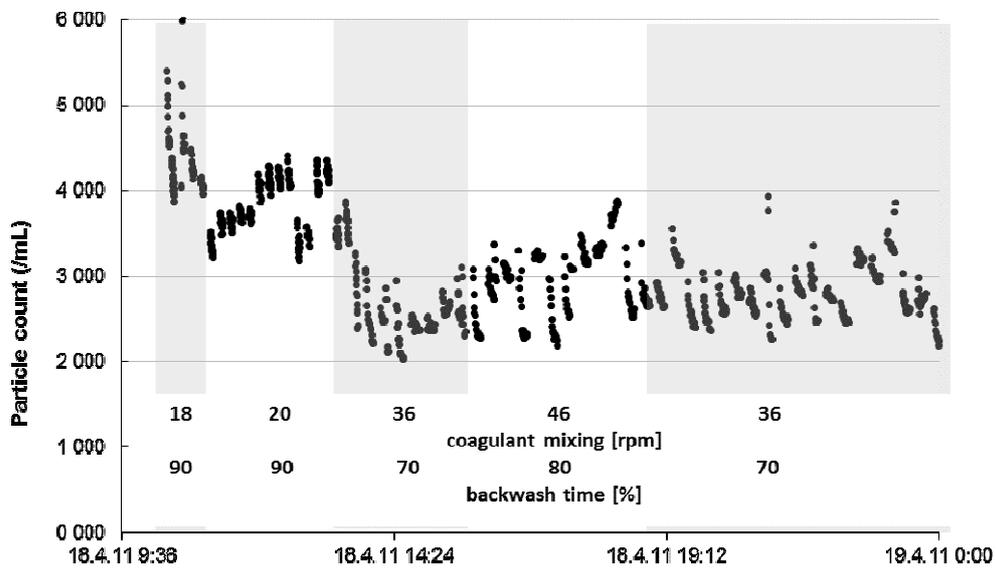


Figure 4: Increase of the mixing velocity in the coagulation tank

Role of organic substances in tertiary treatment via oxidation and membrane filtration

In this work package the influence of different treatments (ozonation, coagulation) on macromolecular organic substances (biopolymers) in secondary effluent and the effects on subsequent ultrafiltration were investigated at lab-scale. Furthermore, fouling mechanisms were intensively investigated and an analytical method was developed to observe the formation of ozonation by-products.

Analyses with LC-OCD showed a significant reduction of major organic foulants (biopolymers) for coagulation while ozonation appeared to transform macromolecules into compounds smaller than approx. 50 nm, see Figure 5. With ultrafiltration tests (PES membranes) it could be shown that coagulation is capable to reduce total fouling resistance to some extent and additional ozonation can further enhance the membrane filtration process. However, ozonation as a pretreatment step caused more irreversible fouling. The lowest irreversible fouling was achieved with coagulation. LC-OCD analyses showed that the transformation of organic matter by ozonation is mainly responsible for the observed increased irreversible fouling of ultrafiltration membranes. Tests with different membranes showed comparable results for pretreated secondary effluent concerning total fouling resistance. In contrast to the observations with all tested UF membranes, for the tested microfiltration membranes irreversible fouling was reduced with additional ozonation. In general, the pore size seems to be strongly influencing irreversible fouling if ozonation is used for pretreatment of membrane filtration.

Intensive investigations of fouling mechanisms using filtration laws identified cake filtration as the dominant filtration process for coagulation while additional ozonation leads to increased pore blocking or in pore fouling, respectively.

Experiments with secondary effluents from different sewage treatment plants in Berlin showed comparable fouling behavior for all observed pretreatments. Thus membrane filtration results generated with samples from WWTP Ruhleben seem to be transferable to other WWTPs in Berlin.

MALDI-TOF-MS analyses of secondary effluent were not suitable to identify major organic foulants, neither in solution nor on top of the membrane after filtration. Consequently, MALDI-TOF-MS was primarily used for investigations of theoretical aspects of fouling by using model fouling substances.

An analytical procedure for bromate was successfully developed with LC-MS/MS at TUB. With the procedure it was possible to quantify samples up to a limit of quantification of 0.5 µg bromate per liter. Higher concentrations of bromate (> 10 µg/L) were produced only at specific ozone consumptions higher than 0.9 mgO₃/mgDOC₀, see Figure 6.

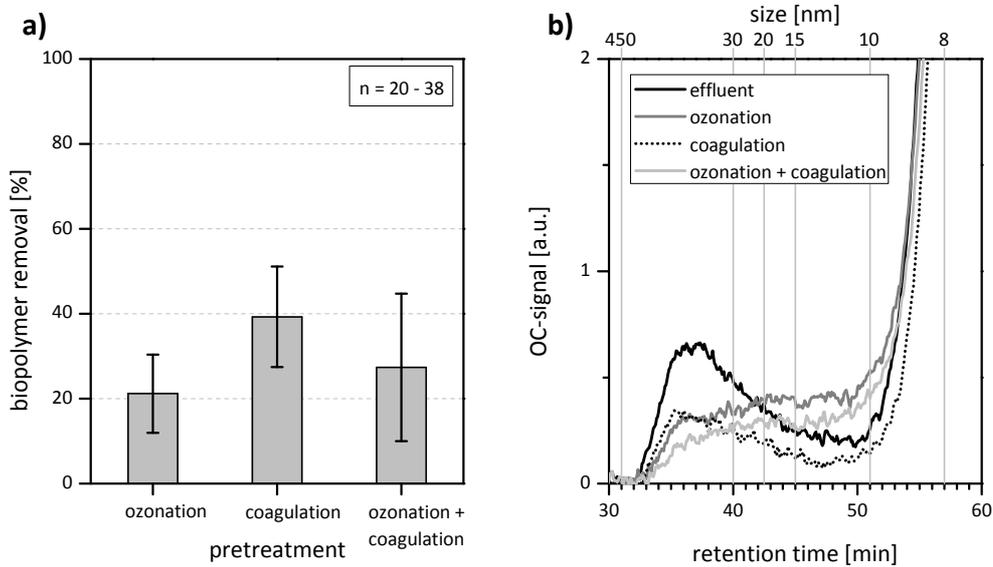


Figure 5: Removal and transformation of biopolymers by different pretreatments; a) removal (mean values with standard deviation for all pretreatment experiments carried out during project runtime) of biopolymers by ozonation ($Z_{spez} = 0.4 - 1.6 \text{ mgO}_3/\text{DOC}_0$), by coagulation ($0.036 - 0.216 \text{ mmol Me}_3^+/\text{L}$), by combination of pre-ozonation and coagulation ($Z_{spez} = 0.4 - 1.6 \text{ mgO}_3/\text{DOC}_0$ and $0.036 - 0.216 \text{ mmol Me}_3^+/\text{L}$) and b) exemplary LC-OCD chromatograms (HW55S column, focus on biopolymers) for the pretreatments.

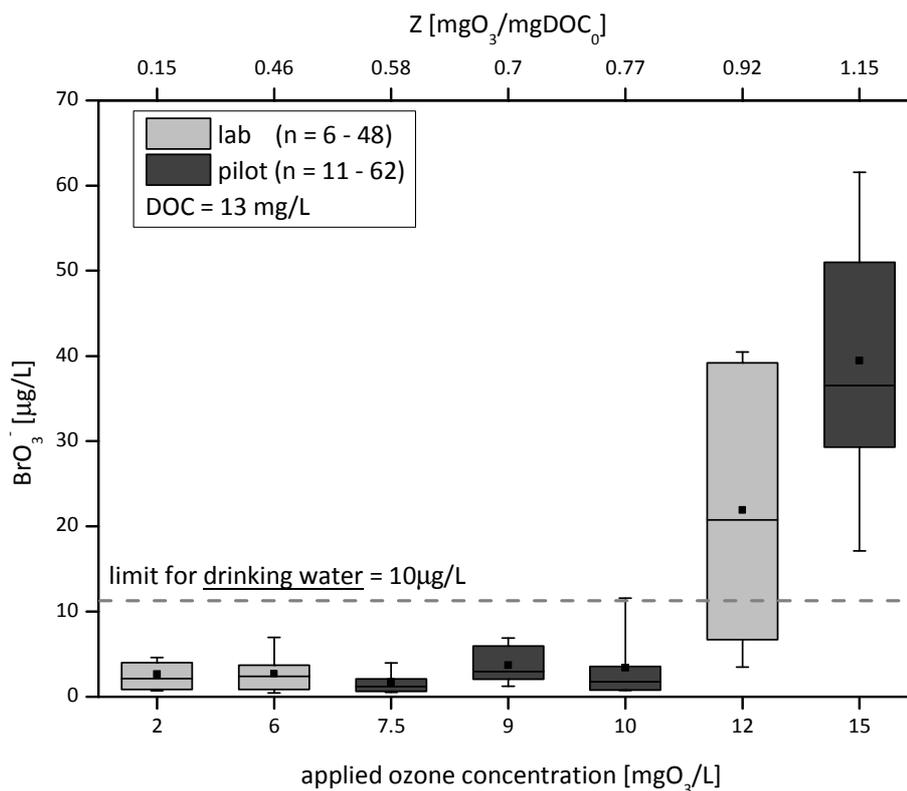


Figure 6: Bromate formation during the experimental phase in the lab and at the pilot plants for different ozone consumptions; experimental phase from March 2011 – December 2012; specific ozone consumption is calculated for $\text{DOC} = 13 \text{ mg/L}$.

Feasibility of the microsieve technology for advanced phosphorus removal

The pilot trials (throughput of 10–30 m³/h) at WWTP Ruhleben proved that the microsieve technology (company Hydrotech, Sweden) combined with chemical pre-treatment achieves good and reliable phosphorus removal with effluent values < 80 µg/L TP, see Figure 7. The first three months of pilot operation confirmed the general process performance observed during the pre-trials in 2009 but also revealed a need for process optimization with regard to the removal of suspended solids and the reduction of coagulant breakthrough. An improved performance was achieved through change from ferric chloride (FeCl₃) to polyaluminum chloride (PACl). In the presented case, PACl gave clearly better results for the removal of phosphorus and suspended solids than FeCl₃. Additionally, the occurrence of coagulant residues could be noticeably reduced. In contrast to FeCl₃, dosing PACl led to an improvement of the water transmittance simplifying disinfection with UV irradiation.

Load proportional dosing of PACl and polymer was introduced in order to avoid under as well as over dosing of the chemicals. The dose of cationic polymer had a significant impact on water quality and backwash time: With the initial process configuration 1.5 to 2 mg/L cationic polymer were recommended for a safe and stable operation with adequate backwash time resulting in an average polymer dose of 1.7 mg/L. However, latest results showed that a polymer dose of only 0.6 mg/L is possible without losses in water quality and filtration performance when mixing conditions were optimized. During the constructional modifications the hydraulic retention time of the coagulation was reduced from 4 to 1 min at peak flow. Due to the installation of a TurbomixTM short-circuiting could be avoided. Furthermore, the turbulence in the flocculation tank was increased. Despite the noticeable reduction of the hydraulic retention time and the polymer dose the rebuild resulted in improved reduction of suspended solids (2.2 mg/L) and coagulant residues in the microsieve effluent. The operation regime of the chemical treatment prior to the microsieve filtration showed to be a trade-off between the energy demand for mixing and the polymer consumption. Due to the continuous operation over more than 20 months important operational experience was gained with regard to backwash behavior and cleaning intervals. The backwash time mainly correlates with the influent flow (10-30 m³/h), the influent water characteristics and the properties of the formed flocs. Due to progressing fouling of the filter panels chemical cleaning was necessary every 4 to 7 weeks. A shorter cleaning interval (e.g. every 4 weeks) might be beneficial as the backwash time and thus the energy demand could be kept on a lower level. In this application the microsieve produced on average 1.8% of backwash water. The backwash water showed excellent settling properties (SVI << 50 mL/g) and might be easily treated via returning to the primary clarifiers.

The UV disinfection unit as post-treatment after the microsieve was operated with a fluence of 730 J/m². Good disinfection could be provided for a continuous operation of 7 months, see Figure 8. During this period a concentration of less than 100 MPN/100 mL of *E. coli* and *Enterococci* in the effluent of the UV disinfection were achieved.

Overall, the microsieve in combination with dosing of coagulant and polymer is a robust technology with low phosphorus effluent values (< 80 µg/L) and a low energy demand of about 21 Wh/m³ (+ site-specific energy demand for water lifting). Microsieving, together with UV disinfection, can be an option for applications targeting phosphorus removal and disinfection, e.g. effluent polishing for sensitive areas or landscape irrigation.

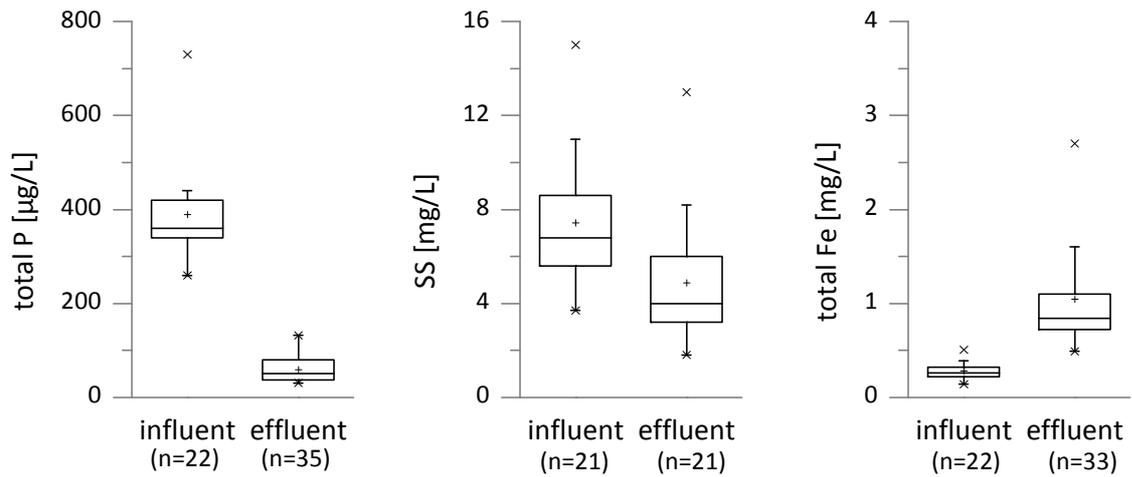


Figure 7: First pilot plant results (October/ November 2010)

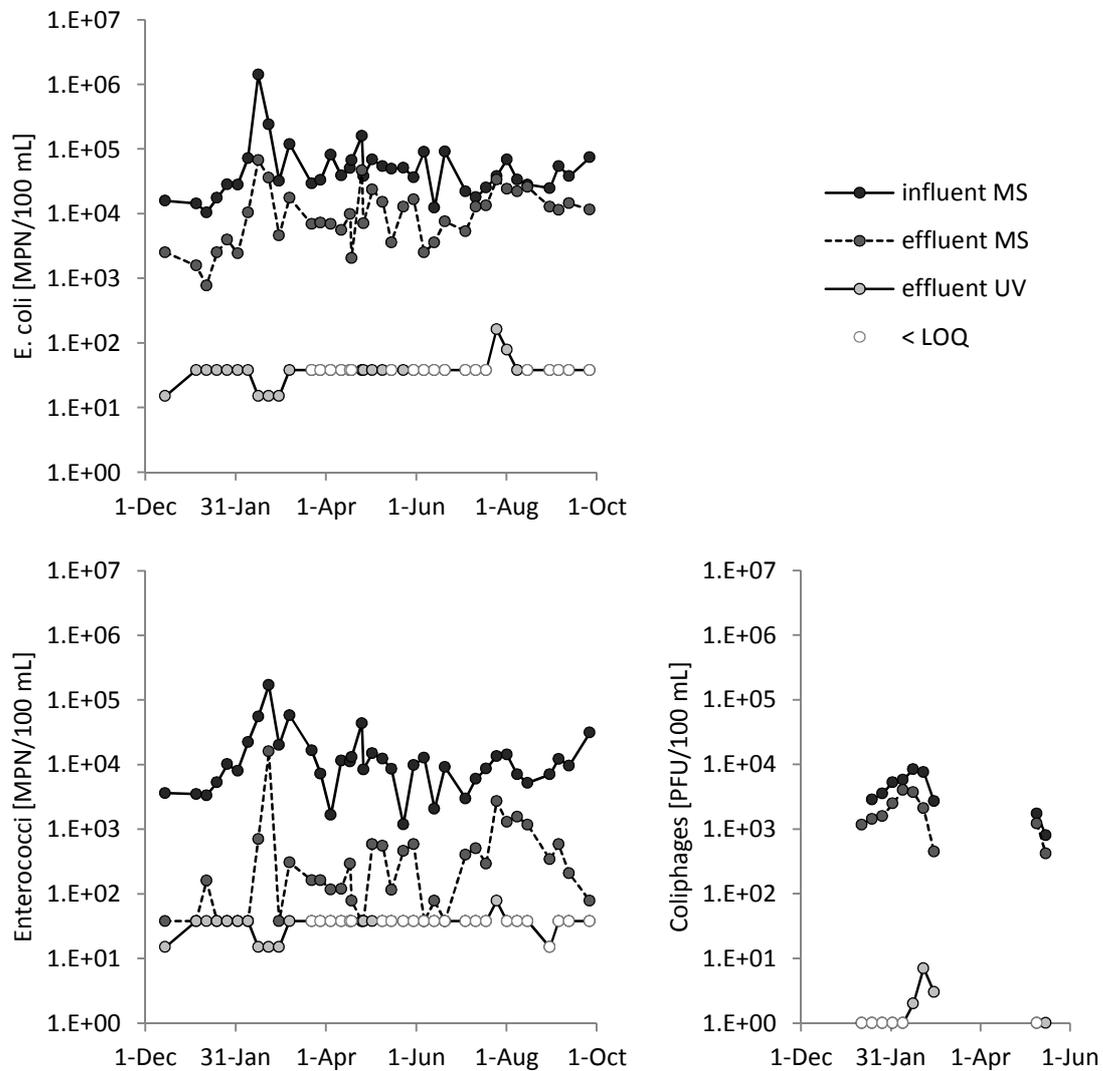


Figure 8: Concentrations of the indicator organisms *E. coli*, Enterococci and Coliphages in the influent and effluent of the microsieve and after the UV disinfection. The limit of quantification (LOQ) for *E. coli* and Enterococci was 15 or 38 MPN/100 mL in dependence of the dilution.

Life Cycle Assessment and Life Cycle Costing of tertiary treatment schemes

For a future upgrade of the wastewater treatment plant (WWTP) Ruhleben targeting advanced removal of total phosphorus (TP) ($< 50\text{-}120\ \mu\text{g/L TP}$) and seasonal disinfection, various technological options for tertiary treatment of secondary effluent are suitable to fulfill these goals. This study applies the holistic methods of Life Cycle Assessment (LCA) and Life Cycle Costing (LCC) to assess and compare those options for tertiary treatment at WWTP Ruhleben in their environmental and economic impacts, including all relevant direct and indirect processes and effects of the WWTP upgrade. Options for tertiary treatment include gravity-driven processes such as dual media filtration (DMF), microsieve filtration (MSF), or high-rate sedimentation (HRS), and membrane-based processes such as ultrafiltration with polymer membranes (Polymer UF) or microfiltration with ceramic membranes (Ceramic MF). For disinfection in the summer period, gravity-driven processes are complemented by downstream UV disinfection, which is only applied in rain weather bypass for membrane processes. Process data for operational parameters and infrastructure design are based on optimized long-term pilot trials at technical scale (DMF, MSF, Polymer UF, Ceramic MF) or conservative process modelling based on supplier information (HRS).

LCA shows that the existing phosphorus load in secondary effluent of WWTP Ruhleben (28 t/a TP) can be reduced substantially by all processes, eliminating 19-25 t/a TP (67-90%) depending on the process, see Figure 9. A minor side-benefit for effluent quality is also expected from the further elimination of heavy metals adsorbed to particulate matter in secondary effluent. At the same time, tertiary treatment schemes will increase energy demand and related emissions of greenhouse gases (carbon footprint) of the existing WWTP process by an estimated 12-21% and 7-13%, respectively. Gravity-driven processes with low coagulant dosing (DMF, MSF, HRS) have a considerably lower energy demand and carbon footprint than membrane-based processes with high electricity demand for feed pumps and higher coagulant dose. At the same time, low-energy treatment processes do not reach the exceptional high effluent quality of membrane-based processes. Consequently, a certain trade-off between energy demand/carbon footprint and effluent quality can be quantified. In analogy to the environmental assessment and effluent quality, LCC results show that total annual costs are lowest for HRS (5.1 Mio €/a) and comparable between DMF and MSF (5.7 Mio €/a), followed by Polymer UF (10.2 Mio €/a) and Ceramic MF (12.2 Mio €/a). In comparison to gravity-driven processes, membrane-based processes are characterized by both higher investment costs (factor 1.5 – 3x) and higher operational costs (factor 2 – 2.5x), mainly due to high costs of membranes, machinery, electricity, and coagulants.

Comparing the relative resource efficiency for selected environmental and economic parameters related to the total load of eliminated phosphorus, DMF and MSF are the most efficient of the assessed technologies for tertiary treatment, spending $\sim 250\ \text{€/kg P}_{\text{elim}}$ and causing $180\ \text{kg CO}_2\text{-eq/kg P}_{\text{elim}}$ (both with UV disinfection as post-treatment). HRS + UV has higher relative costs ($270\ \text{€/kg P}_{\text{elim}}$) and higher carbon footprint ($235\ \text{kg CO}_2\text{-eq/kg P}_{\text{elim}}$) due to the lower effluent quality of the process (= less reduction in TP loads). Membrane-based processes have the highest relative costs for P removal ($400\text{-}475\ \text{€/kg P}_{\text{elim}}$) and the highest carbon footprint ($275\ \text{kg CO}_2\text{-eq/kg P}_{\text{elim}}$): even though their superior effluent quality leads to the highest total reduction in TP loads, the high energy demand and costs of membrane processes yield higher relative spending of resources related to the final goal, see Figure 10.

Based upon the pilot results and the LCA / LCC investigations, a simplified Excel based model was developed that enables to perform pre-planning of advanced phosphorus tertiary treatment (with or without UV disinfection) for any large WWTP. The model

enables to screen cost-efficiency and resource-efficiency of the processes considered in the project Oxeram, and to identify design and operation criteria leading to minimum specific costs and environmental impacts.

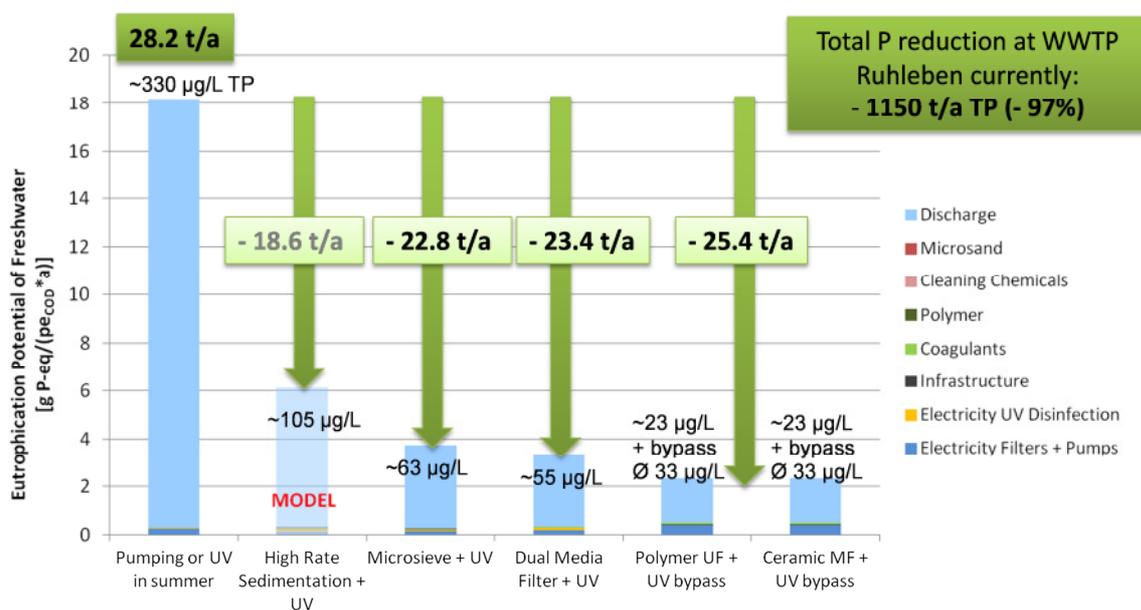


Figure 9: Phosphorus reduction of different schemes for tertiary treatment

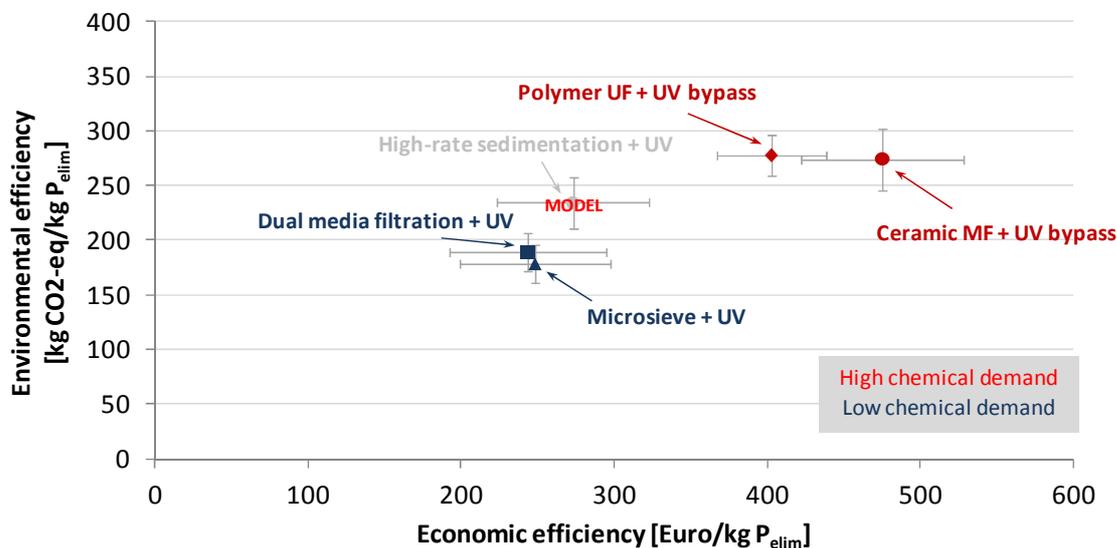


Figure 10: Environmental and economic efficiency of different schemes for tertiary treatment

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